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Focus paper

Generation and preservation of continental crust in the Grenville Orogeny



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ABSTRACT

Detrital zircons from modern sediments display an episodic temporal distribution of U–Pb crystallization ages forming a series of ‘peaks’ and ‘troughs’. The peaks are interpreted to represent either periods of enhanced generation of granitic magma perhaps associated with mantle overturn and superplume events, or preferential preservation of continental crust during global collisional orogenesis. The close association of those peaks with the assembly of supercontinents implies a causal relationship between collisional orogenesis and the presence of zircon age peaks. Here these two end-member models (episodic periodicity of increased magmatism versus selective preservation during collisional orogenesis) are assessed using U–Pb, Hf, and O analysis of detrital zircons from sedimentary successions deposited during the ~1.3–1.1 Ga accretionary, ~1.1–0.9 Ga collisional, and < 0.9 Ga extensional collapse phases of the Grenville orogenic cycle in Labrador and Scotland. The pre-collisional, accretionary stage provides a baseline of continental crust present prior to orogenesis and is dominated by Archean and Paleoproterozoic age peaks associated with pre-1300 Ma Laurentian geology. Strata deposited during the Grenville Orogeny display similar Archean and Paleoproterozoic detrital populations along with a series of broad muted peaks from ~1500 to 1100 Ma. However, post-collisional sedimentary successions display a dominant age peak between 1085 and 985 Ma, similar to that observed in modern North American river sediments.

Zircons within the post-orogenic sedimentary successions have progressively lower ϵ_{Hf} and higher $\delta^{18}\text{O}$ values from ~1800 to ~1200 Ma whereupon they have higher ϵ_{Hf} and $\delta^{18}\text{O}$ within the dominant 1085–985 Ma age peak. Furthermore, the Lu–Hf isotopic profile of the Grenville-related age peak is consistent with significant assimilation and contamination by older crustal material. The timing of this dominant age peak coincides with the peak of metamorphism and magmatism associated with the Grenville Orogeny, which is a typical collisional orogenic belt. The change from broad muted age peaks in the syn-orogenic strata to a single peak in the post-orogenic sedimentary successions and in the modern river sediments implies a significant shift in provenance following continental collision. This temporal change in provenance highlights that the source(s), from which detrital zircons within syn-orogenic strata were derived, was no longer available during the later stages of the accretionary and collisional stages of the orogenic cycle. This may reflect some combination of tectonic burial, erosion, or possibly recycling into the mantle by tectonic erosion of the source(s). During continental collision, the incorporated continental crust is isolated from crustal recycling processes operative at subduction margins. This tectonic isolation combined with sedimentary recycling likely controls the presence of the isotopic signature associated with the Grenville Orogeny in the modern Mississippi and Appalachian river sediments. These results imply that zircon age peaks, which developed in conjunction with supercontinents, are the product of selective crustal preservation resulting from collisional orogenesis.

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1. Introduction

Continental crust is a key repository of Earth history, preserving the product of geological processes that have shaped the planet in deep time. This record is vulnerable to destruction through the processes of sediment subduction and subduction erosion at convergent plate margins (Scholl et al., 1980; von Huene and Scholl, 1991; Stern, 1991; Clift et al., 2009; Stern, 2011), and through lower crustal delamination (Bird, 1979; Kay and Mahlburg Kay, 1993; Houseman and Molnar, 1997; Schott and Schmeling, 1998; DeCelles et al., 2009). The locus of crustal recycling is primarily at convergent plate margins where the subducting oceanic slab carries a veneer of sediment and it tectonically erodes crustal material from the overriding plate into the mantle. Paradoxically, it is also along convergent plate margins where the vast majority of continental crust is generated. Scholl and von Huene (2009) postulated that at present the ratio of crustal formation and destruction is roughly balanced, resulting in a zero net gain of continental crustal volume. In contrast, Stern (2011) estimated the total current rate of crustal recycling to be greater than the rate at which the crust is being generated by magmatic activity, and so the present total volume of continental crust may be decreasing.

The degree to which the current distribution of continental crust represents the original volume generated has been considerably debated (e.g. Bowring and Housh, 1995; Hawkesworth et al., 2009, 2010; Condie et al., 2011; Cawood et al., 2013). The temporal heterogeneity of presently exposed continental crust lies at the heart of this issue. There is considerable discussion whether the “peaks” and “troughs” of continental crust formation ages (broadly represented by zircon U–Pb crystallization ages) represent periods of episodically increased generation of continental crust (Condie, 1998; Rino et al., 2004; Yin et al., 2012; Arndt and Davaile, 2013; Walzer and Hendel, 2013) or selective crustal preservation (Hawkesworth et al., 2009; Condie et al., 2011; Roberts, 2012). Furthermore, excluding the Archean (see Ernst, 2009; Cawood et al., 2013; Spencer et al., 2014a), zircon age peaks in global compilations of zircon U–Pb ages appear broadly to correspond with the timing of supercontinent formation (e.g., Condie, 1998, 2000, 2003; Hawkesworth and Kemp, 2006; Kemp et al., 2006; Campbell and Allen, 2008; Voice et al., 2011; Arndt and Davaile, 2013).

Proponents of episodes of enhanced magmatism rely on significant mantle–plume activity or mantle–overturn events (Condie, 1998; Rino et al., 2004; Komiya, 2007; Arndt and Davaile, 2013). However, the andesitic composition of the bulk continental crust (Taylor and McLennan, 1985; Rudnick and Gao, 2003) suggests that near steady-state subduction zone magmatism and the resulting volcanic arcs is the dominant contributor of new continental crust (see McCulloch and Bennett, 1994; Davidson and Arculus, 2006; Hawkesworth and Kemp, 2006; Cawood et al., 2013). Roberts (2012) proposed that the lack of zircons with depleted ε_{Hf} during supercontinent assembly reflects increased crustal recycling during these periods, which he relates to the geodynamic configuration (subduction polarity, age of colliding crust, etc.) of the assembling continental fragments (see also Murphy et al., 2003; Murphy et al., 2009; Collins et al., 2011).

The alternative viewpoint is that the peaks and troughs in the zircon age archive are artifacts of varying preservation potential during and between times of collisional orogenesis leading to the assembly of supercontinents (Hawkesworth et al., 2009; Condie et al., 2011; Roberts, 2012; Cawood et al., 2013). The simplified stages of continental collision display three stages within a generalized geodynamic “cycle” of subduction, collision, and rifting phases (Hawkesworth et al., 2009, Fig. 1). Assuming the longevity of mass balance within modern subduction zones, crustal preservation

during the subduction phase is minimal despite the large volumes of crust forming from subduction zone magmatism. This is especially true for advancing subduction margins (*sensu* Cawood et al., 2009) where large amounts of crust are removed by sediment subduction, subduction erosion, and delamination (Clift et al., 2009; DeCelles et al., 2009; Stern, 2011). The preservation potential increases in retreating continental margins in that slab retreat is greater than the subducting plate velocity resulting in significant intra-arc and back-arc extension and generation of continental crust and in extreme examples, oceanic crust (e.g. Cawood et al., 2009). As collisional orogenesis begins and subduction zone magmatism ceases, the latter stages of subduction zone magmatism are often preserved within the foreland of the collisional orogeny (as in the Transhimalayan volcanic arc; Hodges, 2000). Additionally, magmatism produced during the continental collision is dominated by lower crustal melts forming within an over-thickened crust and compressed thermal gradients along with decompression melting during orogenic exhumation (Harris et al., 1986). A key aspect of the selective preservation model of Hawkesworth et al. (2009) is that the detrital zircons that make up an ‘age peak’ may be largely derived from the latest stage of subduction zone magmatism rather than magmatism solely associated with the collision (Fig. 1), which is a period of minimal new crust generation. Collision is followed by the collapse and eventual rifting of the continental crust and relatively minor volumes of mafic magmatism and crustal generation (Scholl and von Huene, 2009; Cawood et al., 2013). Given the above postulates, it is predicted that detrital zircons derived from a collisional orogen will exhibit a dominant age peak associated with the large-volume of latest-stage subduction related magmatism preserved during continental collision.

These first order tectonic processes and the associated ideas for the growth and loss of continental crust are often assessed using combined isotopic systems (particularly U–Pb, Hf, O) primarily linked to voluminous compilations of isotopic analyses of zircon. It is argued that zircons from large rivers that drain large areas of continental crust provide the best way to obtain an unbiased and representative sample of the continental crust therein (e.g. Iizuka, 2005; Campbell and Allen, 2008; Wang et al., 2009; Iizuka et al., 2010; Wang et al., 2011; Yin et al., 2012; Iizuka et al., 2013). However, Allegre and Rousseau (1984) and Dhuime et al. (2011) have demonstrated that continental erosion results in a distinct bias toward younger lithotectonic domains. Other workers use large global databases with tens of thousands of analyses in an attempt to

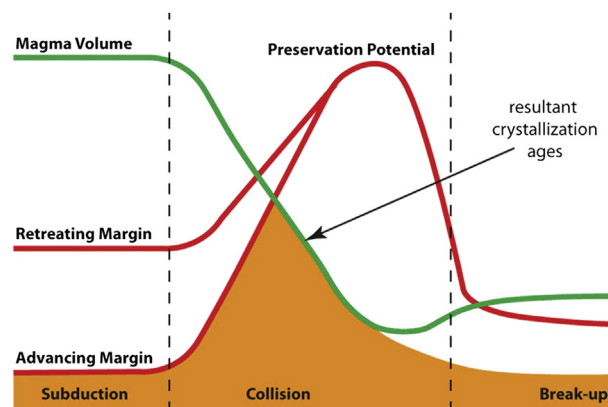


Figure 1. The volumes of magma generated (green line) and their likely preservation potential (red line) vary through the three stages associated with the convergence, assembly, and breakup of a supercontinent (after Hawkesworth et al., 2009). Peaks in igneous crystallization ages that are preserved in the rock record (shaded area) reflect the balance between the magma volumes generated during the orogenic cycle and their respective preservation potential.

“see through the noise” and utilize a variety of modeling parameters to attain a more representative sample of continental crust (Belousova et al., 2010; Condie et al., 2011, 2012; Dhuime et al., 2011, 2012; Voice et al., 2011; Roberts, 2012). Although these methodologies provide a global vantage point of crustal generation and preservation, they are unable to decipher the specifics of individual orogenic systems (as in Kemp et al., 2009; Boekhout et al., 2013; Condie, 2013).

Models that propose that detrital zircon age peaks represent periods of time with increased crustal generation imply a significant influx of isotopically *depleted* material related to island arc and mantle plume magmatism (Stein and Hofmann, 1994; Condie, 1998; Rino et al., 2004; Stein and Ben-Avraham, 2007; Arndt and Davaille, 2013). In contrast, models that interpret zircon age peaks as the product of increased crustal preservation during the assembly of supercontinents predict that they are associated with isotopically *enriched* magmatism due to crustal reworking associated with late stage subduction and collisional orogenesis (Hawkesworth et al., 2009; Condie et al., 2011; Cawood et al., 2013). Further, an additional outstanding issue is at what stage of the orogenic cycle does the age peak develop? This has a significant bearing on whether the zircon age peaks are the product of enhanced magmatism prior to collision (as in Condie, 1998; Rino et al., 2004; Arndt and Davaille, 2013) or increased preservation of the latest subduction-related magmatism during the subsequent collisional stage of the orogenic cycle (as in Hawkesworth et al., 2009).

To test these models of enhanced crustal production or selective crustal preservation, and the timing of zircon peak formation, we dissect the prominent zircon U–Pb age peak within a single discrete orogenic system associated with the assembly of Rodinia. Specifically we focus our study within the environs of the Grenville Orogen, using detrital zircons from a suite of sedimentary rocks in Labrador and Scotland that were deposited before, during, and after the timing of peak orogenesis (Figs. 2–4). Sedimentary successions were selected from key intervals in Labrador and Scotland that can be divided into pre-, syn-, and post-collision timeframes (Figs. 3–5). Detrital zircons from rock units spanning the orogenic cycle allows for the characterization of crustal reworking and recycling during subduction, collision, and rifting, and identification of the timing of dominant age peak development. Coupling U–Pb, Hf, and O isotopic analysis in zircon from these units allows us to investigate: (1) the degree of crustal reworking associated with the respective stages of collisional orogenesis; and (2) how the

prominent age peak seen in modern river sediments is derived from the rock archive linked to the Grenville orogenic cycle.

Hf and O in zircon are particularly useful as they provide a measure of reworking of older continental crust (Hf) along with incorporation of supracrustal material (O). A partial melt of the mantle should have a composition near that of the Hf depleted mantle model (Söderlund et al., 2004). The accumulation of radiogenic ^{176}Hf will drive the $^{176}\text{Hf}/^{177}\text{Hf}$ composition away from the initial ratio based upon the initial $^{176}\text{Lu}/^{177}\text{Hf}$ ratio of the parent magma. Those magmas that in turn assimilate older crustal material will lie along a mixing trend between the depleted mantle and crustal end members. Zircons in equilibrium with pristine mantle-derived melts have $\delta^{18}\text{O}$ values between 4.7 and 5.9‰ (Valley, 2003). Higher $\delta^{18}\text{O}$ values reflect a component enriched in ^{18}O , typically interpreted as resulting from assimilation of supracrustal material into the magma from which the zircon crystallized (Eiler, 2001). Importantly oxygen isotope ratios of igneous rocks are not sensitive to the age of material assimilated (as with Hf isotopes) but they are sensitive to the amount of material that has experienced low-temperature oxygen fractionation (Valley, 2003). Melts that incorporate such sediments therefore have higher zircon $\delta^{18}\text{O}$ values (Valley et al., 1994; Roberts et al., 2013).

2. Geologic setting

Prior to the Grenville Orogeny, the eastern margin of Laurentia (present co-ordinates) consisted of a series of margin-parallel accretionary complexes of late Paleoproterozoic to late Mesoproterozoic age (e.g. 1.8–1.6 Ga Yavapai/Mazatzal/Labradorian, 1.5–1.3 Ga Granite-Rhyolite/Pinwarian provinces, and 1.3–1.1 Ga Composite Arc Belt/Elzevir/Frontenac terranes; see Whitmeyer and Karlstrom, 2007 and Hynes and Rivers, 2010). Together these accreted tectonic zones constitute part of the Great Proterozoic Accretionary Orogen that extended from southwest Laurentia to Baltica (Condie, 2013; Roberts et al., 2013). This protracted period of subduction-related crustal growth and accretion was terminated by the Grenville Orogeny (Fig. 5).

The Grenville Orogeny is a 1085–985 Ma tectonothermal event (as per Gower et al., 2008) resulting from continental collision between the southeastern margin of Laurentia with the north and western margins of the Amazonian and/or Kalahari cratons (Dalziel et al., 2000; Tohver et al., 2002; Jacobs et al., 2008; Ibanez-Mejia et al., 2011) during the assembly of the Rodinian supercontinent (Hoffman, 1991). This orogenic event occurred in two discrete phases, an early phase from 1085 to 1040 Ma, and a later one from 1010 to 985 Ma (Gower et al., 2008). The former is characterized by an initial period of intense crustal thickening and upper amphibolite to granulite facies metamorphism (Darling et al., 2004; Bickford et al., 2008), followed by the intrusion of relatively low-volume granitoid magmatism. The latter phase is best characterized along the parautochthonous northwestern margin of the Grenville Province which occurred during orogenic rejuvenation resulting in upper amphibolite facies metamorphism (Hynes and Rivers, 2010 and references therein). Similar to magmatism during the early phase, the later phase is comprised chiefly of granitoids and pyroxene-bearing granitoids (i.e. Hynes and Rivers, 2010). Following the Grenville Orogeny, extensional collapse and rifting led to the breakup of Rodinia. The initial rifting along this collisional suture is recorded by the opening of the Iapetus Ocean ~620–550 Ma (Williams and Hiscott, 1987; Thomas, 1991; Cawood et al., 2001).

The Sveconorwegian Orogeny is often inferred to be an extension of the Grenville Orogeny (*sensu lato*) (e.g. Gower et al., 1990; Karlstrom et al., 2000; Bingen and Mansfeld, 2002; Cawood et al., 2010), although recent findings have implied geodynamic processes distinct from those in the Grenville Orogeny (Slagstad et al.,

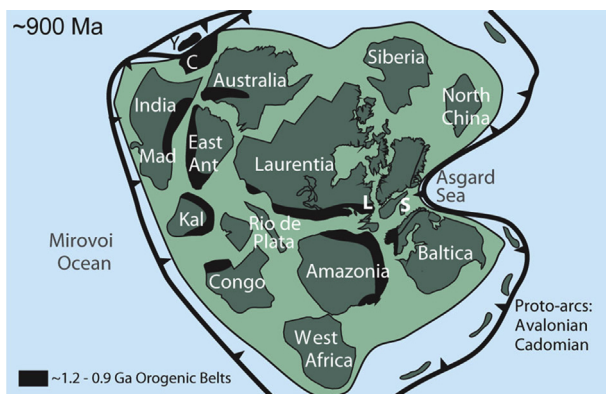


Figure 2. Schematic global paleogeography of Rodinia circa 900 Ma and orogenic events from the preceding 300 My are highlighted (after Gower et al., 2008; Li et al., 2008; Cawood et al., 2010; Cawood et al., 2013). Kal: Kalahari, Ant: Antarctica, Mad: Madagascar, Y: Yangtze, C: Cathysia, L: Labrador, S: Scotland.

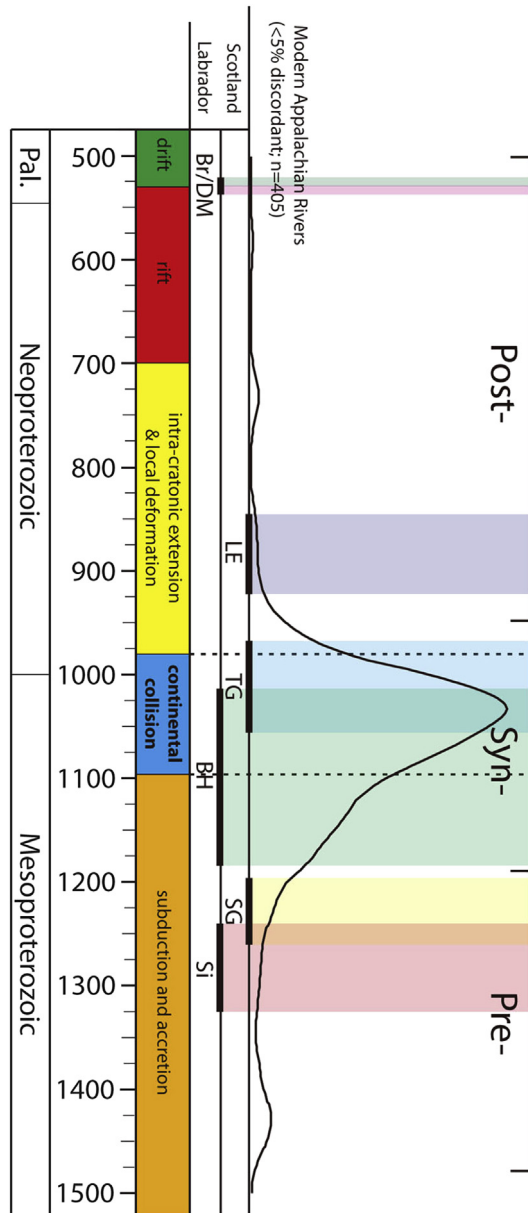
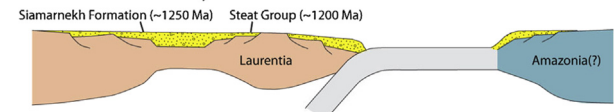


Figure 4. The age range of pre-, syn-, and post-orogenic sedimentary successions sampled in this study overlain by the zircon U–Pb age spectra (kernel density estimation: Vermeesch, 2012) from Eriksson et al. (2012). The vertical axis represents relative probability. Highlighted above the time scale are the stages of the Grenville orogenic cycle (see Section 2 for references). Br: Bradore Formation; DM: Double Mer Formation; LE: Loch Eil Formation; TG: Torridon Group; BH: Battle Harbour Psammite; SG: Sleat Group; Si: Siamarnek Formation.

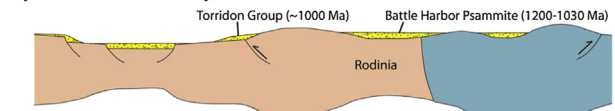
northern Scotland and eastern Greenland (Cawood et al., 2007a, b and references therein; Spencer et al., 2014b). Additionally, detritus from the Grenville Orogen was dispersed across the entire expanse of Laurentia and is preserved in Neoproterozoic sedimentary successions extending to the western and northernmost margins of the craton (Rainbird et al., 1997; Dehler et al., 2010; Rainbird et al., 2012; Spencer et al., 2012).

Although sedimentary rocks from the pre-, syn-, and post-orogenic stages are found throughout much of Laurentia, only eastern Labrador and northern Scotland preserve all three successions, and these regions (Figs. 3 and 4) were targeted for sampling enabling direct comparisons between all three tectonostratigraphic stages.

Pre-collision sedimentary successions:



Syn-collision sedimentary successions:



Post-collision sedimentary successions:

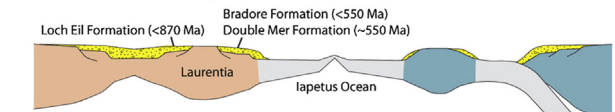


Figure 5. Schematic cross-sections from Laurentia to Amazonia(?) showing positions of sedimentary basins pre-, syn- and post-Rodinian supercontinent cycle (after Cawood et al., 2006). Depositional ages for sedimentary successions are described in Sections 2.2 and 2.3.

2.2. Pre-, syn-, post-orogenic successions in Labrador

The Siamarnek Formation of northern Labrador is composed of subarkose sandstone unconformably overlying the ~1320 Ma Umiakovik Lake batholith (Fig. 3; Wheeler, 1964; Emslie and Loveridge, 1992). The depositional age of the Siamarnek Formation is bracketed by the underlying Umiakovik Lake batholith (Emslie and Loveridge, 1992) and a presumed Grenville-age thrust that cuts the Siamarnek Formation and places the batholith on top of the formation (Wheeler, 1964). The formation is a likely correlative of the ~1.27–1.23 Ga Seal Lake Group ~100 km to the south (van Nostrand and Lowe, 2010). Given the depositional constraints (and the detrital zircon age information described below), this formation is classified as a pre-orogenic sedimentary succession.

The syn-orogenic succession in Labrador is represented by a package of psammitic, semi-pelitic and calc-silicate rocks on Battle Island, near the town of Mary's Harbour (Gower, 2009). Deposition of the supracrustal rocks on Battle Island are constrained between 1200 and 1030 Ma by detrital and igneous zircon U–Pb geochronology (Kamo et al., 2011). These units represent the only known occurrence of post-1.5 Ga sedimentary rocks with a Grenville metamorphic overprint in the eastern interior Grenville province.

Post-orogenic Neoproterozoic to Cambrian sedimentation is recorded in the Double Mer and Bradore formations of Labrador and Newfoundland. These were deposited during continental rifting associated with the breakup of Rodinia (Cawood et al., 2001; Cawood and Nemchin, 2001). Both of these formations have an assumed early Cambrian age, although the absence of Cambrian trace fossils in the Double Mer Formation might imply a late Neoproterozoic age (Gower et al., 1986; Williams and Hiscott, 1987). The Double Mer Formation consists of arkosic sandstone, conglomerate, siltstone, and shale and was deposited in a series of rift basins extending at least 300 km inland from the Labrador coast (Gower et al., 1986). The Bradore Formation has a similar lithology to the Double Mer Formation and is dominated by arkosic sandstone, but it is only found within 30 km of the Labrador Coast (including Belle Isle) and on Newfoundland (Cumming, 1983; Williams and Smyth, 1983; Williams and Hiscott, 1987).

2.3. Pre-, syn-, post-orogenic successions in Scotland

The pre-orogenic sedimentary rocks in Scotland are represented by fluvial-alluvial sandstone, sedimentary breccia, and minor

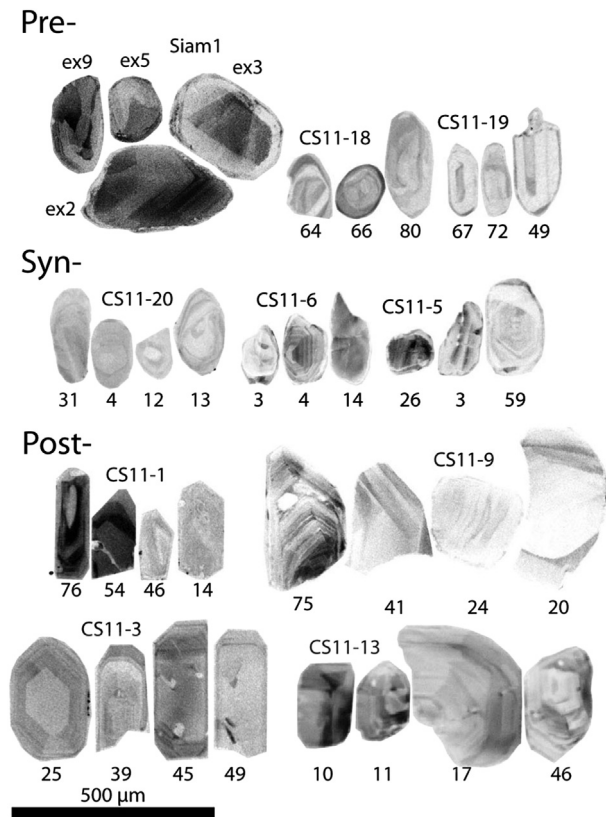


Figure 6. Cathodoluminescence images of representative zircons and in situ locations for O, U-Pb, and Hf analyses from each of the samples in this study.

lacustrine mudstones of the Sleat and Stoer groups (Stewart, 1991; Kinnaird et al., 2007). These groups were deposited in non-marine rift basins and have facies attributed to rapid lateral and vertical changes between coarse- and fine-grained lithologies. They rest

nonconformably on the Paleoproterozoic to Archean age Lewisian basement. Maximum depositional age for the Sleat Group is constrained by the youngest zircon U-Pb date of 1247 ± 34 Ma (Kinnaird et al., 2007). The Stoer Group only contains Paleoproterozoic and Archean zircons (Kinnaird et al., 2007; Lancaster et al., 2011; Williams and Foden, 2011) but a minimum depositional age is estimated using a Pb-Pb isochron age of 1199 ± 70 Ma on a thin stromatolitic bed near the base (Turnbull et al., 1996) as well as an Ar-Ar age of 1177 ± 5 Ma on the impact-related Stac Fada Member (Parnell et al., 2011). The youngest U-Pb zircon analysis from this study (discussed below) also gave an age of 1198 ± 12 Ma further lending credence to a ~ 1200 Ma depositional age.

Overlying the Sleat and Stoer groups is the Torridon Group, which is composed of the basal conglomerate and finer siliciclastic rocks of the Diabaig Formation in turn overlain by the Applecross, Aultbea, and Cailleach Head formations representing syn-orogenic sediments. The Torridon Group is composed of ~ 6 km of upward-fining, fluvial pebbly arkose to siltstone with striking stratigraphic monotony and lithologic homogeneity across hundreds of square kilometers (Kinnaird et al., 2007). Paleocurrent and U-Pb detrital zircon data imply these sediments were derived predominantly from the Laurentian continent to the west (Stewart, 1991; Williams, 2001). Several workers propose that the Torridon Group was deposited in a series of extensional basins that formed during the late-stages of the Grenville Orogeny (Soper and England, 1995; Williams and Foden, 2011). The clockwise rotation of Baltica (between ~ 1120 and 1000 Ma; Salminen et al., 2009; Cawood et al., 2010) and the accompanying opening of the Asgard Sea (between Greenland and Baltica, Fig. 2) facilitated post-Grenville convergence during the Renlandian Orogeny. Cawood et al. (2010) further proposed that sedimentation associated with the Torridon Group was derived from the Grenvillian orogenic belt and deposited within extensional basins resulting from the opening of the Asgard Sea. Other workers, though, consider the facies and stratigraphic characteristics of the Torridon rocks to be more apropos to deposition in the foreland basin of the Grenville Orogeny (Nicholson, 1993; Rainbird et al., 2001; Kinnaird

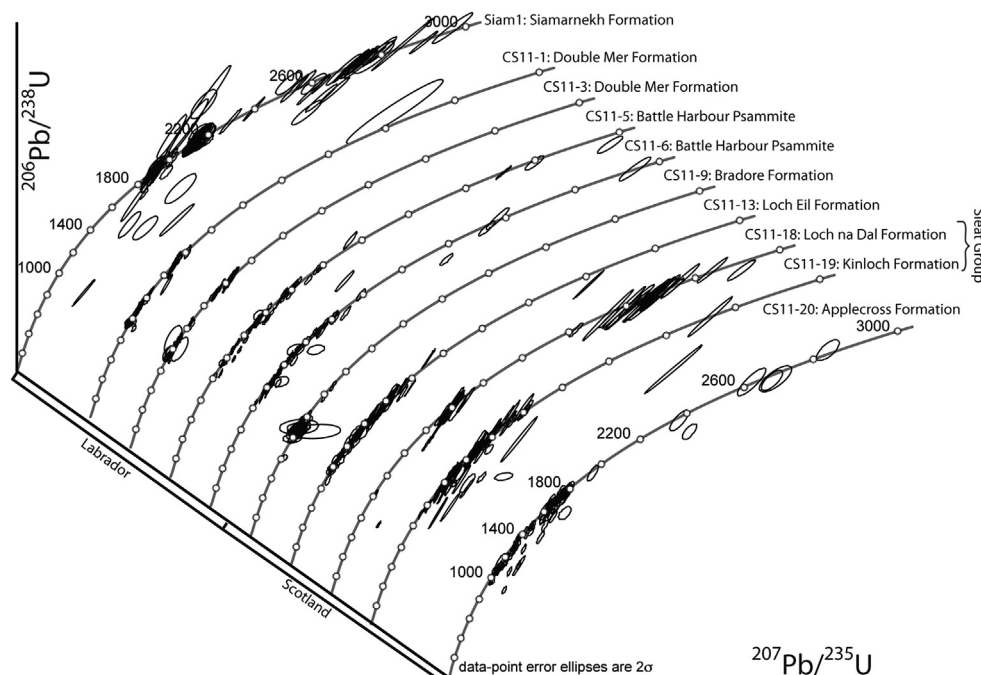


Figure 7. Pb/U concordia diagram of ages (Ma) of zircon grains from each sample. Uncertainties are shown at the 2σ level. Diagram was constructed with the use of Isoplot (Ludwig, 2009).

et al., 2007; Krabbendam et al., 2008). Maximum depositional age of these syn- (to late) orogenic sedimentary rocks is constrained by the youngest U-Pb zircon date of 1046 ± 26 Ma within the upper Torridon Group (Rainbird et al., 2001). The minimum depositional age of 977 ± 38 Ma is obtained using Rb/Sr within a mudstone of the Applecross Formation that presumably dates early diagenesis (Turnbull et al., 1996).

The earliest phase of post-orogenic sedimentation in Scotland is represented by the Morar, Glenfinnan, Loch Eil groups (Soper et al., 1998). These groups form a conformable series of shallow marine sandstones and siltstones interpreted to have been deposited in northeast trending half grabens (Strachan, 1985; Soper et al., 1998). These groups were derived from Laurentia and have maximum depositional ages of ~ 980 Ma (Morar Group), ~ 950 – 900 Ma (Glenfinnan and Loch Eil groups) (Cawood et al., 2004, 2014) and before the intrusion of the West Highland granitic gneiss at 870 ± 30 Ma (Rogers et al., 2001) and the associated metagabbros at 873 ± 6 Ma (Millar, 1999; Cawood et al., 2014).

3. Methods

Ten ~ 3 kg sedimentary samples were collected within each of the pre-, syn-, and post-orogenic successions from localities in south eastern Labrador (Battle Harbor, Double Mer, and Bradore formations) and northern Scotland (Loch Eil, Applecross, and Sleat formations) with a sample from the Siamarnek Formation of northern Labrador. GPS locations of samples are presented in an online KMZ file (Appendix A). Zircons were extracted using standard techniques (i.e. Wilfley table, heavy liquid, Franz magnetic separation), mounted in epoxy resin and polished to expose a cross section through the center of the grains.

Zircons were examined for zoning patterns using cathodoluminescence (CL) and back-scattered electron (BSE) images (Fig. 6). Zircon O isotopic analyses were performed using the Cameca 1270 and 1280 at the Edinburgh Ion Microprobe and NordSIM facilities. Zircon U-Pb and Hf isotopic analyses were performed by laser ablation inductively coupled plasma mass spectrometry (LA-MC-ICP-MS) at the NERC Isotope Geosciences Laboratory, Keyworth, UK (NIGL). Analytical methods and isotopic data are reported in Appendix B.

4. Results

4.1. U-Pb geochronology

Results and instrumentation parameters of U-Pb geochronology are presented in Figs. 7 and 8, and Appendix B. Cathodoluminescence imaging of the detrital zircons shows compositional zoning that is variably complex but is dominated by normal magmatic zonation (Fig. 6). All U-Pb age data is filtered for 5% discordance.

4.1.1. Pre-orogenic successions

Zircons from sample Siam1 display major age peaks at 2150 Ma and 1900 Ma with a few Archean analyses ($^{207}\text{Pb}/^{206}\text{Pb}$ age). Although only 50 of the 129 analyses were $<5\%$ discordant, all analyses have the same age distributions as the concordant subset. Two samples from the Sleat Group were analyzed (Loch na Dal and Kinloch formations). Zircons from the Kinloch Formation have dominant age peaks at 2700 and 1800 Ma. The Loch na Dal Formation has a dominant age peak at 1660 Ma and a spread of ages from 1500 to 1300 Ma ($^{207}\text{Pb}/^{206}\text{Pb}$ age). $\sim 80\%$ of the analyses from the Sleat Group are $<5\%$ discordant.

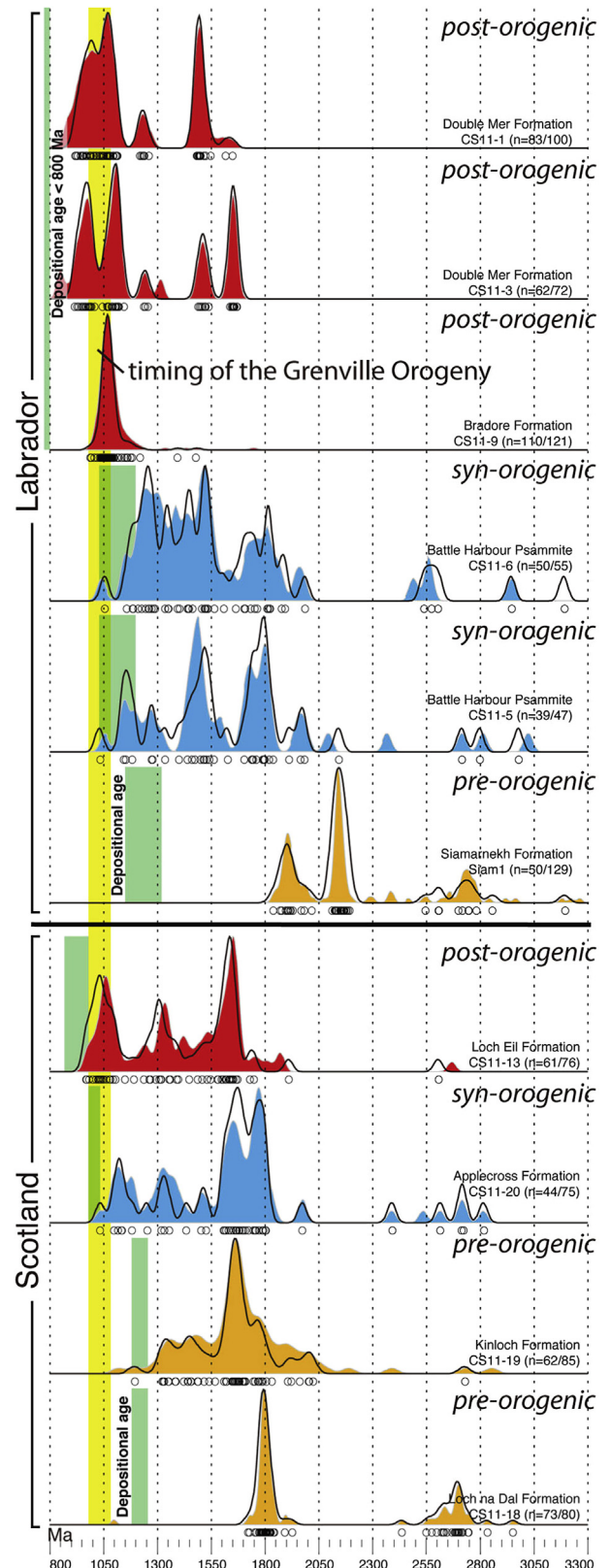


Figure 8. Kernel density estimation (solid line) plots of detrital zircon ages from each sample. Only ages that $< 5\%$ discordant are used. The filled line represents all analyses regardless of discordance. Depositional ages and timing of the Grenville Orogeny are discussed in the Sections 2.2 and 2.3. Plot is constructed using densityplotter (Vermeesch, 2012).

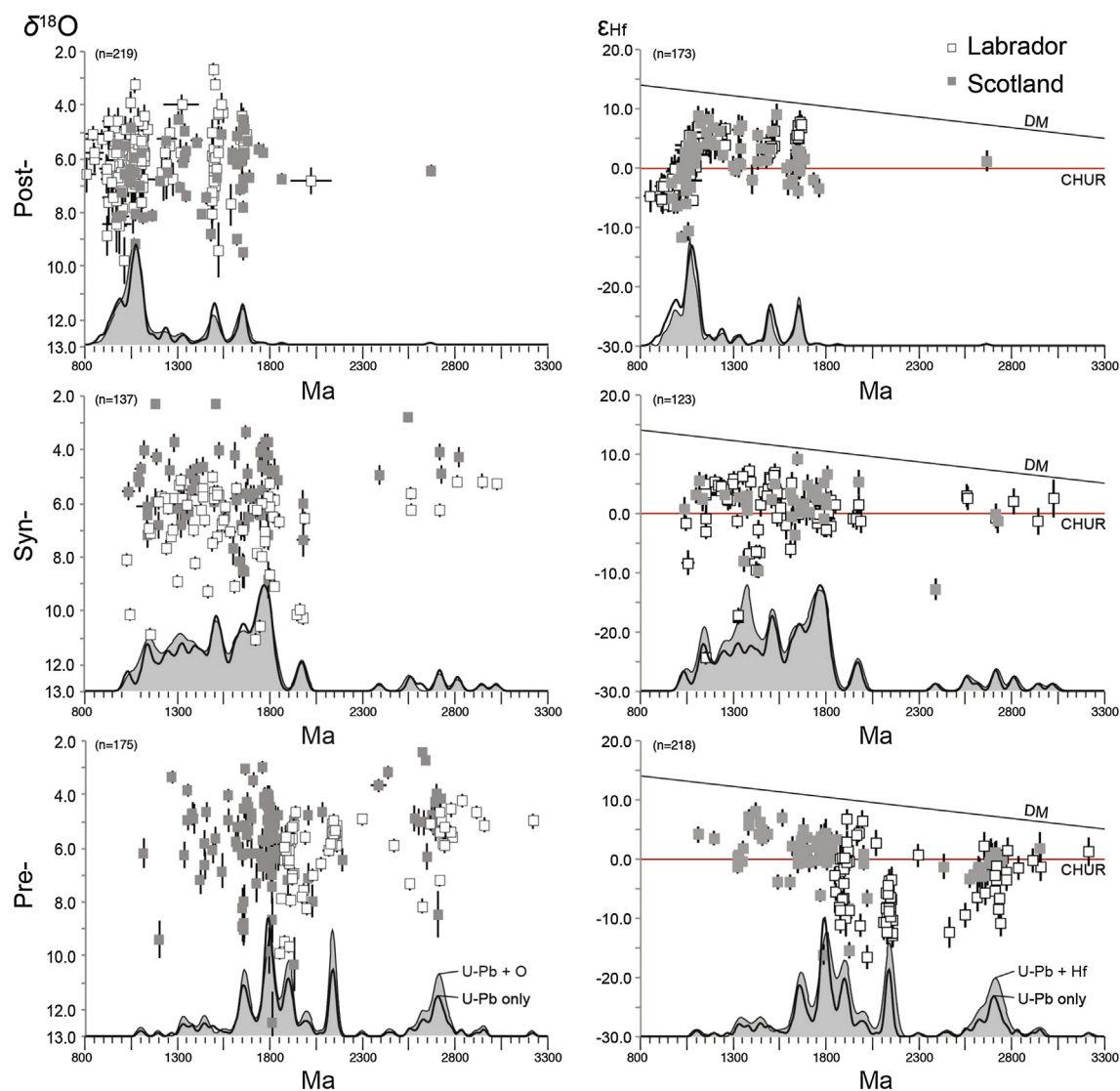


Figure 9. Left: $\delta^{18}\text{O}$ (‰ VSMOW) vs. U-Pb analyses of detrital zircons from the pre-, syn-, and post-orogenic sedimentary successions from Labrador and Scotland. Uncertainties are displayed as 2σ . Right: $\epsilon_{\text{Hf}}(t)$ vs. U-Pb analyses of detrital zircons from the same. Uncertainties are displayed as 2σ . DM: depleted mantle; CHUR: Chondritic Uniform Reservoir.

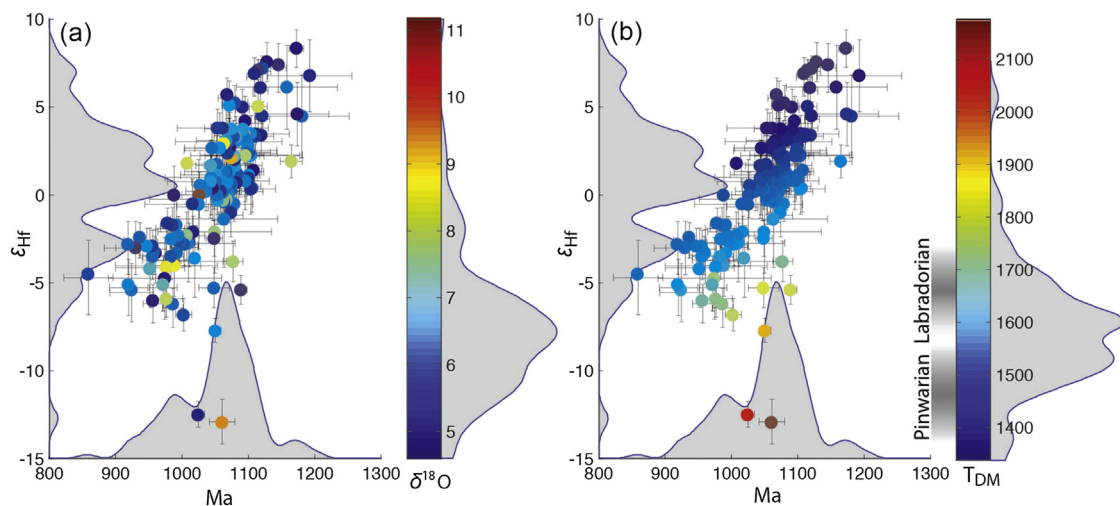


Figure 10. U-Pb age versus ϵ_{Hf} of post-1.2 Ga zircons within the post-orogenic sedimentary successions overlain in (a), with a color spectrum representing $\delta^{18}\text{O}$ values and depleted mantle model ages (T_{DM}) in (b). Along the x-y axes and the color bar in both (a) and (b) are the KDEs of the U-Pb ages, ϵ_{Hf} values, $\delta^{18}\text{O}$ values (‰ VSMOW), and T_{DM} (Ma) respectively. Uncertainties are displayed for both U-Pb ages and ϵ_{Hf} values at 2σ .

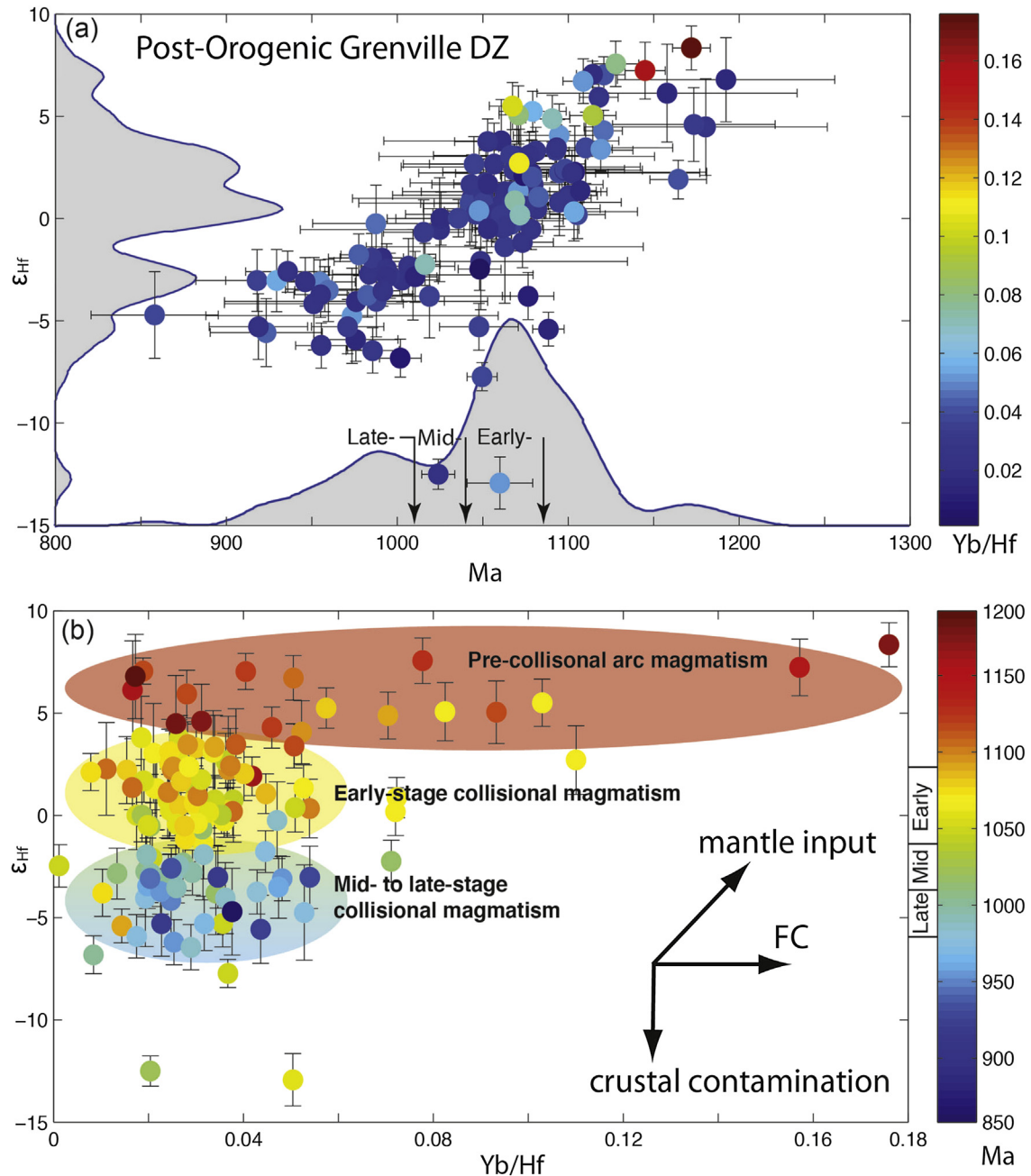


Figure 11. (a) U-Pb age versus ϵ_{Hf} of post 1.2 Ga zircons from the post-orogenic sedimentary successions overlain with a color spectrum representing the measured Yb/Hf ratio in each zircon. Timings of the early and late Grenville Orogeny are after Gower et al. (2008). (b) is same as in (a), although measured Yb/Hf ratio plotted against ϵ_{Hf} with the U-Pb age in the color spectrum. The color spectrum is divided into late-stage subduction zone magmatism, and early-/late-stage magmatism based upon Gower et al. (2008). FC: Fractional crystallization. Trajectories are explained in Section 5.1.

4.1.2. Syn-orogenic successions

Zircons from samples CS11-5 and -6 have dominant age peaks at 1800, 1520, 1250, 1150 Ma and scattered Archean ages ($^{207}\text{Pb}/^{206}\text{Pb}$ age). Nearly 85% of the analyses are < 5% discordant. Zircons from samples CS11-20 show dominant age peaks at 1780 and 1660 Ma, several small peaks between 1550 and 1050 Ma, and several Archean ages ($^{207}\text{Pb}/^{206}\text{Pb}$ age). Although only 44 of the 75 analyses were < 5% discordant, all analyses have the same age distributions as the concordant subset. The combined U-Pb analyses for the syn-orogenic sedimentary successions have a single dominant age peak at 1780 Ma with several sub-peaks of decreasing proportions from 1780 to ~1100 Ma.

4.1.3. Post-orogenic successions

Two samples of the Double Mer Formation were analyzed: CS11-1 has dominant age peaks at 1500, 1230, 1060, and 1000 Ma, and CS11-3 has similar dominant age peaks at 1650, 1500, 1230, 1100, and 980 Ma. ~85% of the analyses are < 5%. Sample CS11-9 has a single dominant age peak at 1060 Ma and 90% of the analyses are < 5% discordant. Zircons from samples CS11-13 have dominant age peaks at 1640, 1300, and 1040 Ma (< 5% discordant subset of $^{207}\text{Pb}/^{206}\text{Pb}$ ages). 80% of the analyses are < 5% discordant. Combined U-Pb analyses for post-orogenic sedimentary successions reveal a single dominant age peak at 1060 Ma and two minor peaks at 1450 and 1600 Ma. In summary, U-Pb zircon age spectra within

the pre-orogenic sedimentary successions are dominated by pre-1.5 Ga ages, the syn-orogenic strata display similar Paleoproterozoic age peaks and a suite of subdued age peaks from 1.5 to 1.1 Ga, and post-orogenic strata are dominated by post-1.1 Ga zircon ages along with minor amounts of pre-1.1 Ga zircon ages (Figs. 7 and 8).

4.2. Zircon Hf and O isotopes

The U-Pb age spectra of each sample and the subset analyzed for Hf and O are nearly identical thereby providing a dataset representative of the U-Pb age spectra (Fig. 9). Pre-orogenic samples have ϵ_{Hf} values that span from the depleted mantle to -15 in pre-1800 Ma zircons and are increasingly depleted to ~CHUR post-1800 Ma. Syn-orogenic samples have a wide spread of ϵ_{Hf} values, mostly between 10 and -10. ϵ_{Hf} values in post-orogenic samples increase from ~CHUR at 1800 Ma to ~5 at 1200 Ma (~0.7 epsilon units per million years) and then decrease from ~5 at 1200 Ma to -6 at 900 Ma (~4.6 epsilon units per million years).

$\delta^{18}\text{O}$ values for pre-orogenic samples range from ~3 to 12‰ and increase from ~3000 to 1800 Ma and decrease thereafter. Syn-orogenic samples have a broad range of $\delta^{18}\text{O}$ values from 2 to 11‰ and post-orogenic samples have a similar pattern to the ϵ_{Hf} values where the range of values decreases from ~1800 to 1250 Ma and then increases thereafter.

5. Discussion

5.1. Zircon age spectra and isotopic composition

U-Pb ages and Hf-O isotopic compositions reported in this study are consistent with those reported previously from the sampled units in Scotland and the Battle Harbour Psammite in Labrador (U-Pb: Friend et al., 1997; Cawood and Nemchin, 2001; Cawood et al., 2007a, b; Kinnaid et al., 2007; Kirkland et al., 2008; Kamo et al., 2011; Hf-O: Lancaster et al., 2011). The depositional provenance of pre-orogenic sedimentary rocks in proximity to the Grenville Orogen (sensu stricto, e.g., Siamarnek Formation and Sleat Group) is characterized by derivation from nearby Laurentian basement (see Gower and Tucker, 1994; Kinnaid et al., 2007). The range of Hf isotopes display increasingly radiogenically enriched values from the ~3.0 Ga zircons until ~1.8 Ga, after which ϵ_{Hf} values become increasingly depleted in post-1.8 Ga zircons (Fig. 9).

Syn-orogenic strata display similar proportions of Mesoproterozoic and Archean zircons. The absence of a dominant Grenville age peak is noteworthy given the syn-Grenville depositional age. For example, the Battle Harbour Psammite and Applecross Formation have depositional ages from ~1200 to ~1030 Ma (Kamo et al., 2011) and ~1200 to ~1050 Ma (Rainbird et al., 2001; Kinnaid et al., 2007), respectively. Assuming a Laurentian derivation, the origin of these Mesoproterozoic zircons can be ascribed to four temporally distinct tectono-magmatic events all of which are related to long-lived convergence from ~1.5 to 1.1 Ga, viz. 1.5–1.3 Ga island arc, continental arc, and back-arc magmatism, 1.3–1.2 Ga continental arc and back-arc magmatism, ~1.1 Ga within-plate magmatism, and 1.09–0.95 Ga magmatism directly associated with collisional orogenesis (see Gower and Korgh, 2002; Hynes and Rivers, 2010). Zircons that formed during each of the distinct periods associated with convergence and collision are nearly equally represented in the syn-orogenic sedimentary successions perhaps signifying that neither a preservation nor production bias is present in these samples; rather the absence of a dominant age peak implies near steady-state zircon production and preservation from ~1.5 to ~1.0 Ga. Hf and O isotopes in zircon from syn-orogenic strata show a wide array of ϵ_{Hf} values for post-1.5 Ga zircons ranging from the

depleted mantle to -25. Similarly $\delta^{18}\text{O}$ values span from ~2 to ~11‰ in post-1.5 Ga zircons. The increasing of $\delta^{18}\text{O}$ and ϵ_{Hf} in these syn-orogenic sedimentary rocks imply significant reworking of supracrustal material inherited from pre-orogenic apparently Paleoproterozoic continental crust since the average depleted mantle model age for post-1.5 Ga zircons is 1.85 Ga.

The dominant Grenville age peak observed in modern/Phanerozoic sediments and global compilations (e.g. Campbell and Allen, 2008; Park et al., 2010; Voice et al., 2011; Eriksson et al., 2012) is only observed in the post-orogenic and not the syn-orogenic sediments analysed. This age peak ranges from ~1150 to 900 Ma and is centered at ~1075 Ma with a younger subpeak at ~990 Ma. These successions display two additional peaks at ~1650 and 1500 Ma, and, in contrast to the syn-orogenic sedimentary successions, less than 10% of zircons analyzed from the post-orogenic successions fall between the ~1500 and ~1075 Ma age peaks ($n = 21$ of 282) and these form two small peaks at ~1240 Ma ($n = 13$) and ~1330 Ma ($n = 8$).

Two discrete age peaks related to the Grenville Orogeny are seen in the samples of the Double Mer Formation. The earlier Grenville peak (ca. 1060–1040 Ma), also seen in the Loch Eil Formation of Scotland, correlates with the earliest phase of the Grenville

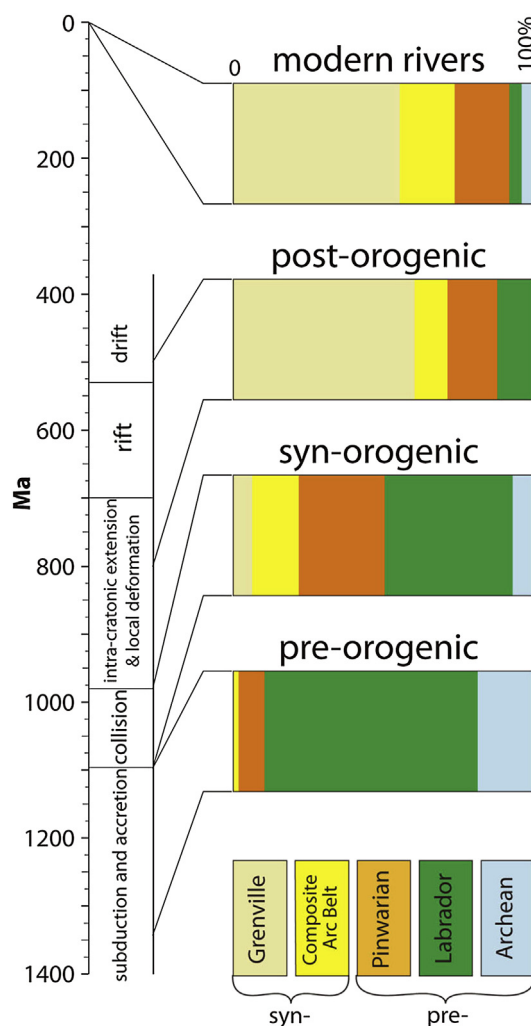


Figure 12. Normalized proportions of the five dominant detrital zircon populations (Archean, Labradorian, Pinwarian, Composite Arc Belt, Grenvillian) within the pre-, syn-, and post-orogenic sedimentary successions and the modern river sediments (Mississippi and Appalachian rivers; data from Izuka, 2005; Wang et al., 2009; Eriksson et al., 2012).

Orogeny (1085–1040 Ma) whereas the younger peak seen in the Double Mer Formation (1000–980 Ma) coincides with the later phase (1010–950 Ma) (phases of Grenville Orogeny after Gower et al., 2008 and references therein). The explanation why the younger age peak is only present in the Double Mer Formation is unclear, for it cannot be attributed to delayed exposure of late-stage magmatic rocks as the Bradore Formation is likely the youngest of the post-orogenic sedimentary successions in Labrador.

Hf and O isotopes in zircon from post-orogenic sedimentary successions display a similar pattern wherein ϵ_{Hf} values become more depleted and the range of $\delta^{18}\text{O}$ values become more restricted near mantle values from ~1800 to ~1200 Ma and vice-versa post-1200 Ma (Fig. 9). The pre-1200 Ma pattern can be attributed either to a decrease in reworking of continental crustal or an influx of isotopically depleted, mantle-derived magma until the initiation of collisional orogenesis wherein crustal reworking resumes.

The post-1200 Ma zircons from post-orogenic formations are examined in detail in Figs. 10 and 11. Fig. 10a displays the U-Pb age against the ϵ_{Hf} value with $\delta^{18}\text{O}$ values plotted using a color spectrum for each analysis. From this, no systematic pattern can be observed between the U-Pb age, ϵ_{Hf} , and $\delta^{18}\text{O}$. This implies that, although the range of $\delta^{18}\text{O}$ values increases post-1200 Ma, there is no direct connection between the degree of radiogenic Hf and ^{18}O enrichment (i.e. increasing depleted mantle model age and supracrustal reworking, respectively). In Fig. 10b, depleted mantle model ages are plotted along a color spectrum against the U-Pb age and ϵ_{Hf} values for each analysis. Depleted mantle model ages generally

increase through time with the highest frequency between ~1.65 and 1.5 Ga. This trend likely reflects the assimilation of a greater proportion of older crustal material through time.

Yb/Hf ratios provide additional insight into the geochemical transition from the latest stages of subduction-related magmatism (1215–1110 Ma; Chiarenzelli et al., 2010; Mclelland et al., 2010) to the early and late collisional phases of the Grenville Orogeny (Gower et al., 2008). The partition coefficients of Yb and Hf into zircon are sufficiently different (Fujimaki, 1986) to show differential element fractionation during fractional crystallization, crustal assimilation, and magma chamber rejuvenation. During fractional crystallization Hf is preferentially incorporated into zircons driving the Yb/Hf ratio of the parent magma up without affecting the ϵ_{Hf} value, whereas assimilation of older crustal material will result in lower Hf isotope ratio (decreasing ϵ_{Hf}) leaving the Yb/Hf ratio constant. An influx of depleted mantle material will likewise leave the Yb/Hf ratio mostly unaffected, but will increase the ϵ_{Hf} values. In general, the Yb/Hf ratios and ϵ_{Hf} values of the pre-collision zircons exhibit a pattern consistent with fractional crystallization within a magmatic system (Fig. 11). Zircons attributed to syn-collisional magmatic systems (1.2–1.1 Ga) show little variation in the Yb/Hf ratios and a significant shift in ϵ_{Hf} values during the collisional phases. Additionally, the younger subpeak seen in the U-Pb spectra in post-orogenic sedimentary successions (Fig. 10a) is likely associated with the later phase of the Grenville orogeny is also apparent in the ϵ_{Hf} spectra. This implies a shift in the composition of material assimilated into the younger magmatic system (Fig. 10).

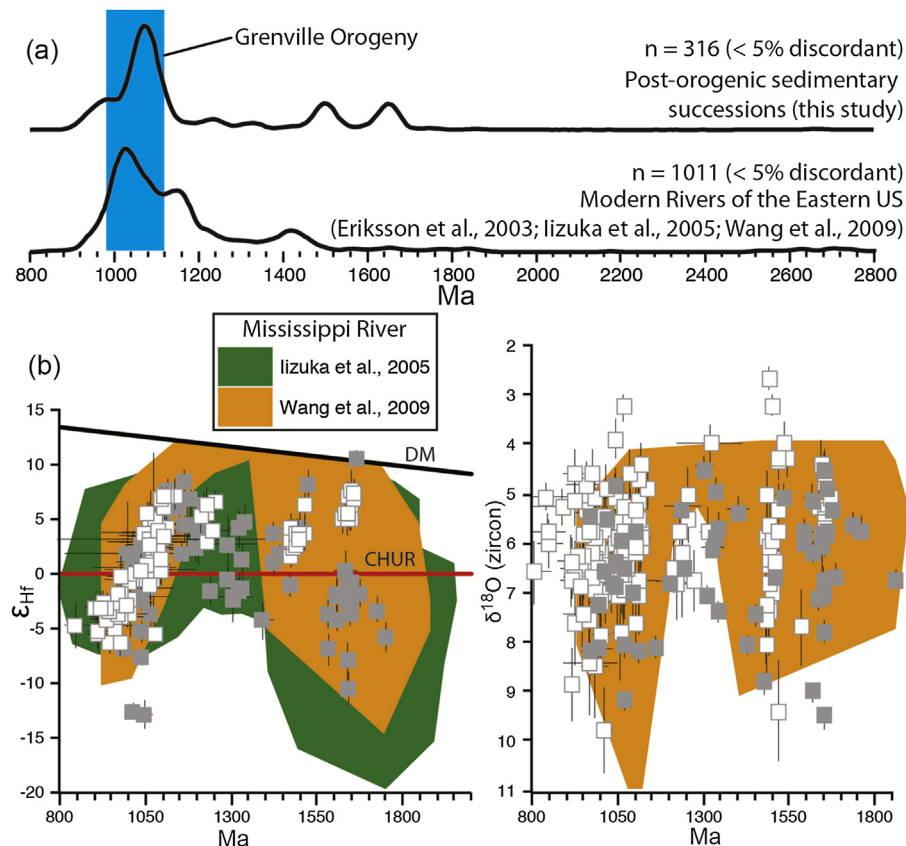


Figure 13. (a) KDE of zircon age spectra of the combined post-orogenic sedimentary successions from Labrador and Scotland (top) compared with the KDE of zircons ages from modern rivers draining the Appalachian mountains (data from lizuka, 2005; Wang et al., 2009; Eriksson et al., 2012). (b) U-Pb ages versus ϵ_{Hf} (left) and U-Pb ages versus $\delta^{18}\text{O}$ (‰ VSMOW) (right) of the post-orogenic sedimentary successions from Labrador and Scotland (symbols are the same as in Fig. 9). Underlying fields represent ϵ_{Hf} and $\delta^{18}\text{O}$ compositions in zircons from the modern Mississippi river delta (data from lizuka, 2005 and Wang et al., 2009). Each of the datasets shows similar 'mustache' patterns wherein values become increasingly depleted in radiogenic Hf (increasing ϵ_{Hf}) and in ^{18}O (decreasing $\delta^{18}\text{O}$; note that $\delta^{18}\text{O}$ axis is flipped) implying greater influx of increasingly mantle-like material (ϵ_{Hf}) with lesser degrees of supracrustal recycling ($\delta^{18}\text{O}$) until post-1.2 Ga whereupon zircons have a more isotopically enriched chemical signature.

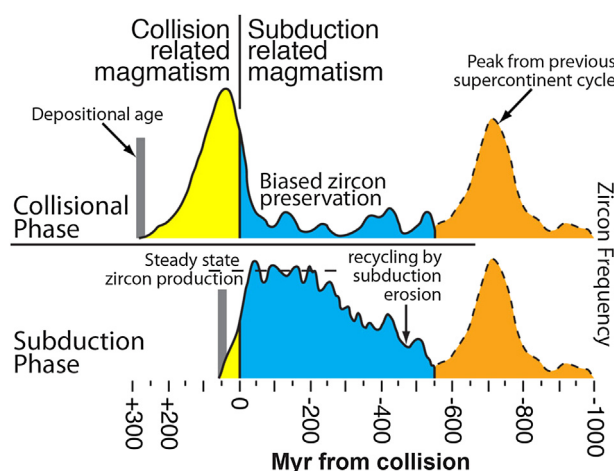


Figure 14. The idealized zircon age spectra during the subduction phase displays the age peak associated with the previous supercontinent cycle along with a broad plateau of ages representing steady-state zircon growth during subduction-related magmatism. Subduction erosion continually removes the oldest zircons associated with the phase of the orogenic cycle. This process continues until continental collision (and supercontinent assembly) continues to remove the previously formed subduction zone-derived zircons. Zircons that are formed during collision-related magmatism are preserved within the interior of the recently collided continents thus protecting it from future subduction erosion. This allows for significant temporal perpetuation of the age peak associated with the collisional orogenesis and the assembly of a supercontinent.

5.2. Increased preservation potential?

The U-Pb age spectra of the pre-, syn-, and post-Grenvillian sedimentary successions display a significant shift in the proportions of zircons of different ages (Fig. 12). This shift is interpreted to represent a significant shift in the crustal provinces available as sources to the sedimentary record. Strikingly, the modern Mississippi and Appalachian rivers (Iizuka, 2005; Wang et al., 2009; Eriksson et al., 2012) display similar detrital zircon population age distributions to the post-orogenic sedimentary successions, implying the crustal availability of Grenvillian detritus has remained relatively constant since the end of the Grenville Orogeny. Moreover, the ϵ_{Hf} and $\delta^{18}\text{O}$ values of detrital zircons within the Mississippi River display the same pattern as seen in the post-orogenic sedimentary successions (Fig. 13b). It is important to note the modern drainages discussed here are separated from the depositional sites of the Bradore, Double Mer, and Loch Eil formations by thousands of kilometers (Fig. 12). Despite this, the similarities in zircon isotopic patterns from modern and Neoproterozoic sedimentary rocks imply that tectonomagmatic provinces from which these zircons are derived display a peculiar isotopic longevity from their initial formation and preservation in the Laurentian craton to their modern levels of exposure and erosion. Rainbird et al. (2012) showed that the Grenvillian zircon age peak is also present throughout the western margin of Laurentia from the Grand Canyon in the south to the Mackenzie Mountains in the north. These far-flung regions with a dominant Grenvillian age signature imply detritus initially shed from the Grenville orogenic belt was transported ~4000 km across Laurentia (see also Rainbird et al., 1992; Rainbird et al., 1997; Dehler et al., 2010; Gehrels et al., 2011; Spencer et al., 2012). The key observation from the data summarized on Fig. 12 is that detrital zircons with relatively old ages, as seen in the pre- and syn-orogenic sediments, do not survive long in the geological record whereas zircons formed in the collisional Grenville orogeny were locked into the upper crust to remain a key feature of the detrital record from 1 Ga to the present day. The residence times of the Grenville aged zircons in the sedimentary record would appear to be longer than those in the syn-Grenville sedimentary successions.

The steady state production of zircon during the subduction phase of the orogenic cycle and consequent recycling of subduction-related magmatism is displayed schematically in Fig. 14. The subduction phase is characterized by significant crustal recycling of subduction-related magmatic rocks back into the mantle. This is in contrast to magmatism that occurs during the collisional phase, which is isolated within the interior of the colliding continents. Continental collision protects and isolates the collision-related tectonomagmatic belt from the various tectonic processes responsible for recycling of continental crust back to the mantle along the continental margins (i.e. mountain root foundering/delamination, tectonic erosion) (Fig. 14). This tectonic isolation of collision-related magmatic rocks leads to a longevity of the detrital zircon isotopic signature associated with the collisional phase of the orogenic cycle. Therefore, the zircon age peak associated with the Grenville Orogeny seen in the modern river sediments of North America is best explained by selective preservation of continental crust during collisional orogenesis (see Hawkesworth et al., 2009). The age peak associated with the Grenville Orogeny is concurrent with the timing of the magmatism and peak metamorphism associated with collisional orogenesis, rather than a period of subduction zone magmatism. This is manifest through the increasingly negative ϵ_{Hf} and greater $\delta^{18}\text{O}$ values from the onset of collisional orogenesis. Alternative models that propose an influx of juvenile, isotopically depleted magmatism are inconsistent these observations (Condie, 1998; Rino et al., 2004; Komiya, 2007; Arndt and Davaille, 2013).

The zircon age peak associated with the supercontinent of Rodinia is one of several dominant zircon age peaks found in global compilations of detrital zircon ages in modern sediments (Campbell and Allen, 2008; Hawkesworth et al., 2010; Iizuka et al., 2010; Voice et al., 2011). As noted by Spencer et al. (2013), variation in geodynamic configuration (subduction zone polarity, advancing/retreating continental margins, longevity of subduction zone magmatism, thickness of leading margin sediments, etc.) can dramatically affect the resulting isotopic signature of a particular age peak and potentially the proportions of latest stage subduction-related magmatism versus collision-related magmatism. Despite the variations in geodynamic styles associated with the individual zircon age peaks this study implies that zircon age peaks, which developed in conjunction with supercontinents, are the product of selective crustal preservation during collisional orogenesis.

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Appendix 1. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.gsf.2014.12.001>.

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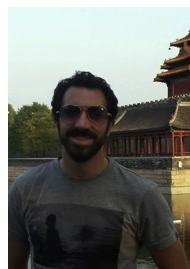
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